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BASIC PRINCIPLES GOVERNING THE FORMATION OF HIGHLY STABLE CONCENTRATED EMULSIONS

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[A Digest]

The factors governing the stability of concentrated emulsions are stable protective films, small drop size, and the ability of the drops to execute Brownian movement. As we showed earlier (1935 - 1936), in a highly concentrated emulsion, when most drops are one micron in diameter, high stability depends upon the formation of structure due to the proximity of the strongly deformed drops. Our laboratory studies on highly concentrated, gelatinized emulsions, e.g., 265 cubic centimeters of benzol in one centimeter^3 of 5-percent sodium oleate solution, show that a tough film covering can maintain stability for many months and that any disturbance of the cell walls or honeycombs causes the emulsion to break down. Such emulsions can be stabilized by protecting the layers with a critical thickness of 0.01-micron sodium oleate solution, which has low resistance to breakdown. Such an emulsion, therefore, is easily broken down by shaking with an excess of organic liquid. Shaking with several drops of water, however, not only prevents breakdown of the emulsion but also re-establishes the emulsion because addition of water increases the distance between the drops. If a greater amount of water is added, a dilute emulsion is obtained.

According to Rebinder's work in 1926 - 1930 and 1930 - 1938, the conditions necessary for stability in concentrated emulsions are adequate surface activity of soluble emulsifiers and stability of the adsorption layer. But if there is a considerable number of large drops without Brownian movement, uniform distribution is disturbed and interphase layers appear, due to insufficient kinetic stability.

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Kinetic stability can be compensated for even if there is a small distance between the drops; under the circumstances, the layer walls of the structure must be staong enough to support the drops, according to Talmud and Bresler (1934).

The present state of colloid chemistry permits the construction of Talmud and Bresler's so-called "spumoid" structures in dilute emulsion in such a way that the movement of drops is impeded. To bring about a sudden increase in a concentrated emulsion's stability, thixotropic colloidal solutions of surface-active stabilizers, which are easily reformed after cessation of mechanical action, can possibly be used. For that purpose the stabilizer should have: (1) surface activity relative to water-oil interphase and adequate surface strength and (2) a capacity for rapid formation of thixotropic systems with rapid restoration of the spatial structure following emulsification.

Thixotropic solutions without noticeable surface activity are not effective stabilizers (e.g., iron hydroxide or vanadium pentoxide), since shaking causes the cells to break and the drops to coalesce. Only thixotropic solutions of surface-active substances can cover drops with a protective layer, even when the network between the drops is disrupted by shaking the emulsion. Hence, although emulsification must necessarily first disrupt the spatial structures in the stabilizing solution, separation into layers should be prevented by instantaneous and rapid restoration of the disrupted structure after cessation of mechanical emulsification.

An emulsion becomes kinetically stable for long periods if the structural absorption layers on the dispersed drops are rapidly connected by structural free-solution fragments during repeated gelatinization. The structure should be easily disturbed by mechanical emulsification in order to overcome resistance without using large energies.

These systems should be studied and characterized by measuring the viscosity of solutions at various pressures and flow velocities. We employed such studies to obtain very stable emulsions by using thixotropic solutions. In this connection we shall briefly describe our experiments with the stabilizing properties of sodium oleate with aliphatic alcohol to obtain concentrated emulsions:

Benzol was the dispersed phase. The alcohol was mixed either with benzol or a one-percent soap solution. Viscosity was measured by Ostwald's viscosimeter in Kroyt's apparatus; surface tension, by Rebinder's maximum-drop and bubble-pressure method. Emulsions were made by stirring stabilizer solutions with benzol by means of a 400-rpm agitator for 5 minutes. Stability was determined by the emulsion's rate of layer formation in graduated cylinders.

Our research showed that alcohol in benzol or soap solution generally increases the stability of oil-water type emulsions. Stability is higher, (1) the higher the quantity of alcohol of any chain length, and (2) the longer the chain for equal alcohol concentrations. The spatial structures of solutions containing sodium oleate and the middle members of the homologous series of aliphatic alcohols are rather fragile, but the emulsion's stability is considerably increased. Cetyl alcohol, however, brings an exceptionally abrupt increase in stability when used in benzol or soap solution. In fact, as small a quantity as 0.1 percent of cetyl alcohol in a one-percent sodium oleate solution is sufficient to form an emulsion which is practically nonseparating for long periods. With increasing alcohol content, the emulsion becomes stable without limit and nonseparating for a year or longer if stored in tightly closed containers. In this connection, we cannot understand Shulman and Cockbain's reference to the very poor stability of emulsions prepared with the aid of sodium oleate and containing cetyl alcohol in the oil phase. Their attempt at a theoretical proof of this is also not clear.

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We obtained viscometric results on soap solutions as a function of cetyl-alcohol content, in the form of curves of relative viscosity at 20 degrees centigrade plotted against pressure. Our data showed that in a sodium oleate solution, cetyl alcohol forms spatial structures which are stronger the higher the alcohol concentration. Thus, we can see that the spatial structures developed in the solution are partly disturbed at increased flow rates but are rapidly re-established during states of rest. In effect, for alcohol concentrations up to one percent, the spatial structure is fully restored almost immediately after disruption brought about by forcing the solution through fine capillaries. With less than 0.1 percent of alcohol, structure formation is not observed in soap solutions; also, these solutions do not show any increase in stability of the emulsion. Solutions weaker than 0.06 percent do not "structurize" and do not influence stability.

Temperature has been shown to influence structure formation, which increases with decrease in temperature.

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